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Cocrystallization through the use of a salt: The case of thiourea with a new propanediammonium oxalate salt

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ABSTRACT

A new blue photoluminescent 3D supramolecular salt namely 1, 3 propanediammonium oxalate has been synthesized and characterized by FT-IR spectroscopy and powder/single crystal X-ray diffraction. The incorporation of the environmental harmful thiourea was investigated. Single crystals were obtained after incorporation and the X-ray diffraction revealed the self-assembly of thiourea with 1, 3 propanediammonium oxalate. The thiourea interacts with the salt through strong hydrogen bonds. The intensity of the blue luminescence slightly decreases upon incorporation of thiourea.

1. Introduction

For the design of supramolecular organic salts, alkylammonium cations are interesting building blocks. Primary alkylammonium can create hydrogen bonds through the three hydrogen atoms therein. A counterion such as oxalate (four acceptors of hydrogen bonds) can be used with such cations and lead to supramolecular salts. Moreover, Carboxylate ions such as oxalate incorporated in film were shown early in the literature acting as luminescent centers [1-3]. Luminescent materials may be useful for photoluminescence sensing.

Photoluminescence sensing is a contact-less and non-destructive method and has been widely used for the detection of some harmful materials such as highly toxic metals or environmental pollutants [4-5]. It was also used for the recognition of small molecules/solvents by incorporating them into luminescent materials [6-9]. Photoluminescence sensing may be therefore useful for the detection of environmental harmful molecules such as thiourea.

Thiourea may be found at elevated concentrations in water or air, due to the multiple uses of this chemical in photography, dyes, drugs, flame retardants, or pesticides [10-14]. It was shown that this molecule has a very slow biodegradability [10] and elevated concentrations are toxic to human, and microorganisms [10]. Several methods have been reported for the determination of trace amounts of thiourea by Raman spectroscopy [15], spectrophotometry [16], titrimetry using iodine [17], high performance liquid chromatography (HPLC) [18] electrochemical methods etc. [19-25]. However, most of these methods have drawbacks such as instrumental

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three-dimensional supramolecular salt, namely 1, 3 propanediammonium oxalate resulting from the mixture of low cost reagents 1, 3 propanediamine and oxalic acid. We successfully incorporated the thiourea into this new luminescent salt.

2. Experimental section

2.1 Materials

Oxalic acid (99%), 1, 3 diaminopropane (99%), thiourea (99%), were purchased from Sigma-Aldrich and used without further purification. Distilled water was used in the syntheses and the incorporation of thiourea.

2.2 Synthesis of compound 1

The synthesis was carried out at room temperature. The compound (1) $\text{NH}_3(\text{CH}_2)_3\text{NH}_3\cdot\text{C}_2\text{O}_4$ (1,3 propanediammonium oxalate) was prepared by mixing 1, 3 diaminopropane (4.4g/59mmol) and dihydrated oxalic acid (7.4g/59mmol) in distilled water. Evaporation at 60°C was carried out for one week. Colourless crystals suitable for X-ray diffraction were obtained. The synthesis diagram is presented in Fig. 1.

Fig 1. Synthesis diagram of compound (1) (hydrogen bonds are represented in dotted line).

2.3 Thiourea incorporation

The incorporation was carried out at room temperature. We mixed a portion of the previously synthesized salt (compound 1) (0.56g/3mmol) and thiourea (0.2g/3mmol) in distilled water. Evaporation at 60°C was carried out. After three days, colourless crystals suitable for X-ray diffraction were obtained. The synthesis diagram is presented in Fig. 2.

Fig. 2. Synthesis diagram of compound (2) (hydrogen bonds are represented in dotted line).

2.4. X-ray diffraction

Single-crystal X-ray diffraction for the two compounds was performed using an Xcalibur, Atlas, Gemini ultra diffractometer. For the refinement, in the absence of significant anomalous scattering, Friedel pairs were merged. The absolute configuration was arbitrarily assigned.

Table 1. Crystal data and structure refinement for compounds 1 and 2

Compound	1	2
Molecular formula	$C_2O_4 \cdot C_3H_{12}N_2$	$2(C_3H_{12}N_2) \cdot 2(C_2O_4) \cdot CH_4N_2S$
Formula weight	164.17	404.44
Crystal color, habit	Plate, colorless	Plate, colorless
Dimensions, mm	$0.52 \times 0.37 \times 0.16$	$0.49 \times 0.28 \times 0.11$
Crystal system	Orthorhombic,	Triclinic
a , Å	8.8036 (12)	6.6949 (10)
b , Å	9.7527 (15)	8.8020 (11)
c , Å	4.2015 (6)	16.3336 (14)
α , deg	90	76.600 (9)
β , deg	90	79.085 (11)
γ , deg	90	83.429 (11)
V , Å ³	360.74 (9)	916.9 (2)
space group	$Pba2$	$P-1$
Z	2	2
$F(000)$	176	432
θ range, deg	6.5–66.7	3.6–29.5
Absorption coefficient, mm ⁻¹	1.12	0.23
T , K	100	100
λ , Å	1.54184	0.71073
Reflections collected	2594	5938
Independent reflections	638 [R(int)= 0.074]	4561 [R(int)= 0.077]
Final R indices	R1=0.041, wR2=0.109	R1=0.062, wR2 = 0.137
R indices (all data)	R1=0.0424, wR2 = 0.113	R1=0.0849, wR2 = 0.187
GOF on F^2	1.033	0.979
Peak, hole /eÅ ⁻³	-0.19 , -0.33	-0.96–0.66

The relatively large ratio of minimum to maximum corrections applied in the multiscan process (1: nnn) reflect changes in the illuminated volume of the crystal. Changes in illuminated volume were kept to a minimum, and were taken into account [26] by the multi-scan inter-frame scaling [27]. The H atoms were all located in a difference map, but those attached to carbon atoms were repositioned geometrically. The H atoms were initially refined with soft restraints on the bond lengths and angles to regularize their geometry (C—H in the range 0.93–0.98, N—H in the range 0.86–0.89 N—H to 0.86 O—H = 0.82 Å) and $U_{iso}(H)$ (in the range 1.2–1.5 times U_{eq} of the parent atom), after which the positions were refined with riding constraints [28].

Computing detail are the followings: Data collection: CrysAlis PRO 1.171.38.43 (Rigaku OD, 2015); cell refinement: CrysAlis PRO 1.171.38.43 (Rigaku OD, 2015); data reduction: CrysAlis PRO 1.171.38.43 (Rigaku OD,

2015); program(s) used to solve structure: SIR97 (Altomare et al., 1999) [29]; program(s) used to refine structure: CRYSTALS (Betteridge et al., 2003) [30]; molecular graphics: CAMERON (Watkin et al., 1996) [31]; software used to prepare material for publication: CRYSTALS (Betteridge et al., 2003) [30]. The details of the crystal parameters, data collections and refinement for compound 1 and 2 are summarized in Table 1. Selected bonds and angles for the two compounds are available in supporting information (Table S1 and S2). Details about hydrogen bonds for both compounds are listed in supporting information (Table S3 and S4).

Powder X-ray diffraction (PXRD) patterns were recorded for the salt (compound 1) with a Bruker AXS D8 diffractometer with CuK α radiation (λ = 1.5406 Å) at room temperature.

2.5 Spectroscopy

Infra-Red spectroscopy and photoluminescence measurements were carried out on compound (1). IR measurement was performed using ATR (Attenuated Total Reflectance) method [32]. PL properties were characterized by a Spex Fluorolog-3 spectrofluorometer from Jobin Yvon Instruments. Diffuse reflectance spectrum of the compound (1) was also collected with a Perkin Elmer UV-visible-NIR spectrometer (Lambda 1050) equipped with an integrating sphere.

3. Results and discussion

3.1 Crystal structures

The single-crystal X-ray diffraction performed on compound (1) reveals that it crystallizes in an orthorhombic system with space group *Pba*2. The asymmetric unit contains half of the anion and cation. The same measurement performed on compound (2) reveals that it crystallizes in a triclinic system with space group *P*-1. The asymmetric unit contains two units of the cation and the anion of compound (1), and one unit of thiourea.

Oxalate anion has four sites which can act as acceptor of hydrogen bonds. On the other hand, each of the two ammoniums of the 1, 3-propanediammonium (PDA) has three hydrogen atoms available for hydrogen bonding. Therefore, oxalate and PDA are connected via N-H...O-C strong hydrogen bonds ($d_{\text{N-H}\cdots\text{O}}$ is in the range of 1.9-1.95 Å) (see supporting information table S3). Some of O atoms interact with two different cations through bifurcated hydrogen bonds leading to a three-dimensional network.

After mixing thiourea and the previously synthesized salt in water and after evaporation, a resulting material is formed where thiourea was completely incorporated into the supramolecular salt (Fig. 3). The two units interact via N-H...O-C strong hydrogen bonds (see supporting information table S4: $d_{\text{N3-H31}\cdots\text{O7}^i}$; $d_{\text{N3-H32}\cdots\text{O10}}$; $d_{\text{N4-H41}\cdots\text{O16}^{ii}}$). Crystal structure was drawn using Diamond and Mercury software.

Fig. 3. Crystal structure of thiourea incorporated into supramolecular 1, 3 propanediammonium oxalate.

3.2 Powder X-ray diffraction

Powder X-ray diffraction shows that the salt (1) is pure. All the diffraction peaks of the powder can be indexed to the orthorhombic phase with space group *Pba*2 (Fig.4).

Fig. 4. Simulated (100K) and experimental (room temperature) XRD patterns of 1, 3 propanediammonium oxalate.

3.3 FT-IR spectroscopic study

The FT-IR Spectrum of the compound (1) is shown in Fig. 5. The full attributions of the main absorption bands are listed in Table 2. According to the literature [33-36], the broad absorption band in the range of 3018-2519 cm⁻¹ is assigned to ν_{N-H} vibration modes of NH₃ in PDA and ν_{C-H} (CH₂) of alkyl chain. The broadness of this band is due to the strong intramolecular hydrogen bonds N-H...O as observed in the crystal structure. The vibration bands from 1631 to 1520 cm⁻¹ are due to ν_{as}COO⁻ while the ones from 1293 to 1222 cm⁻¹ are assigned to ν_sCOO⁻. ν_{C-N} and ν_{C-C} appear respectively at 1055 and 765 cm⁻¹. Therefore, the presence of the cation (PDA) and the anion (oxalate) is confirmed.

For the compound (2), the FT-IR spectrum is shown in Fig. 6. The full attributions of the main absorption bands are available in Table 3 [33-36]. Both spectra of compound (1) and (2) are very similar. But we can identify the presence of thiourea in addition to the cation and the anion of compound (1). Indeed the previously broad band attributed to ν_{N-H}, ν_{C-H} (CH₂) shifts

to 3274 cm^{-1} , due to the presence of thiourea which induces supplementary $\nu\text{N-H}$ and intramolecular hydrogen bonds $\text{N-H}\cdots\text{O}$. Then, the band assigned to $\nu\text{N-H}$, $\nu\text{C-H}(\text{CH}_2)$ appear from $3274 - 2500\text{ cm}^{-1}$. νasCOO^- appear from $1617\text{-}1500\text{ cm}^{-1}$ while νsCOO^- in the range of $1388\text{-}1293\text{ cm}^{-1}$. The vibration band at 1400 cm^{-1} is attributed to $\nu\text{C=S}$. The ones at 1054 and 760 cm^{-1} are respectively assigned to $\nu\text{C-N}$ and $\nu\text{C-C}$.

The results of the FT-IR measurements for both compound (1) and (2) agree with the crystal structure determined by single crystal X-ray diffraction by confirming the presence of the cation, the anion and strong intramolecular hydrogen bonds $\text{N-H}\cdots\text{O}$.

Fig. 5. FT-IR spectrum of 1, 3 propanediammonium oxalate.

Table 2. Assignment of the FT-IR bands of compound (1).

$\nu(\text{cm}^{-1})$	Assignments
3018 - 2519	$\nu\text{N-H}$; $\nu\text{C-H}(\text{CH}_2)$
1631, 1618, 1546, 1520	νasCOO^-
1293, 1222	νsCOO^-
1055	$\nu\text{C-N}$
765	$\nu\text{C-C}$

Fig. 6. FT-IR spectrum of 1, 3 propanediammonium oxalate with incorporated thiourea.

Table 3. Assignment of the FT-IR bands of compound (2)

$\nu(\text{cm}^{-1})$	Assignments
3274 - 2500	$\nu\text{N-H}$; $\nu\text{C-H}(\text{CH}_2)$
1617, 1570, 1522, 1500,	νasCOO^-
1388, 1293	νsCOO^-
1400	$\nu\text{C=S}$
1054	$\nu\text{C-N}$
760	$\nu\text{C-C}$

3.4 UV-Visible absorption

Diffuse reflectance (Fig. 7) shows absorption from 250 to 400nm for the salt (compound 1). This UV absorption may be assigned to $\pi-\pi^*$ and $n-\pi^*$ transition in oxalate ion. HOMO -LUMO energy gap of oxalic acid was already calculated in the literature to be 5.74 eV ($\lambda = 343\text{ nm}$) [37]. It was shown that after co-crystallization of oxalic acid with the 2-(benzylamino) pyridine (HOMO -LUMO energy gap = 6.66 eV), the resulting material had a significantly reduced energy gap 3.47 eV ($\lambda = 572\text{ nm}$) [37]. The same phenomenon was observed by Faizan *et al.* [34] for another dicarboxylate salt: they observed that the HOMO-LUMO energy gap for the resulting salt was significantly lowered than its constituents; they concluded that this reduction may be due to the proton transfer process ($\text{N}^+-\text{H}\cdots\text{O}^-$) and hydrogen bond formations. This may explain why the absorption of our material is extended below 400nm.

Fig. 7. UV-VIS Reflection spectrum of compound (1).

3.5 Luminescence properties

The emission spectra of oxalate ion via UV region excitation were earlier shown in the literature to be around 470 nm [2, 3, 38, 39]. The presence of σ bond and especially delocalized π bond are responsible for the excitation of oxalate ion in UV region [3]. The mapping photoluminescence of the compound (1) was performed with excitation from 250 to 360 nm and emission from 380 to 700 nm . As shown in Fig. 8, the PL mapping exhibits a broad emission centered at 470 nm . According to the literature [2; 3], this excitation may be attributed to the oxalate ion, the unique optical center of

the material. Owing to the absence of oxidizing agent in our compound, photochemical decomposition such as the one reported by Vitz [40] is unlikely to be responsible of the luminescence". The photoluminescence of the resulting material in which thiourea is incorporated was also investigated. The later material showed similar blue photoluminescence (Figure S1). A slight decrease of the emission intensity was observed. This decrease would originate from the difference of concentration of oxalate (the luminescent centers) which is higher in (1) than in (2).

Fig. 8. PL mapping of the compound (1).

4. Conclusion

A new compound 1, 3 propanediaminium oxalate was synthesized at room temperature by mixing, in distilled water, dihydrated oxalic acid and 1,3 propanediamine in 1/1 molar ratio. The material is blue luminescent and consists of tridimensional network owing to strong N-H $\cdots$ O hydrogen bonds. An investigation on possible incorporation of thiourea in this later material was carried out successfully. Thus, we believe the incorporation of this molecule into a photoluminescent material may be useful for its future detection.

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